Polychlorinated Biphenyl Residues in Milk of Environmentally and Experimentally Contaminated Cows

by G. F. Fries*

Polychlorinated biphenyl (PCB)[†] residues have been found in the milk of cows. In some instances the residue levels exceeded the FDA guideline 0.2 ppm in milk (equivalent to 5.0 ppm in milk, fat), and the milk was removed from the market. The major source of PCB residues in milk is Aroclor 1254 that has been used in coatings for concrete silos. Aroclor 1254 is unaltered in silos, and most of the contamination is adjacent to the walls (1, 2).

We have observed a number of farms with PCB-treated silos and have fed Aroclor 1254 to cows under controlled conditions. This paper summarizes our major findings.

Nature and Occurrence of Residues

The PCB recovered from silage is identical to standard Aroclor 1254 (1). However, many of the components with short gas chromatographic retention times do not occur in milk residues (Fig. 1). The components with short retention times are apparently less chlorinated than those with long retention times. Our observations in cows are similar to observations in other species (3).

A complication in analyzing environmental samples is a possibility of interference by DDT and some of its related compounds. We dehydro-

chlorinated the samples to remove possible interferences of DDD and DDT. DDE did not interfere, because those components of Aroclor 1254 with the same retention time as DDE did not occur in significant amounts in milk (Fig. 1).

We obtained milk tank samples from six farms with PCB-contaminated silos. The PCB levels in milk fat are summarized in Table 1. In all cases PCB concentration in milk fat exceeded the FDA guideline, at least part of the time, when silage was fed. When the silage was not fed, levels of PCB in milk fat were always lower than the guidelines.

The farms represent a variety of feeding and management conditions. The results suggest that any dairyman with a contaminated silo will probably exceed the guideline at least some of the time during the year. While a few higher levels have been reported for herds removed from the market, most were within the range of our observations. The number of farms with PCB-contaminated silos is not known. However, PCB contamination of milk is probably more prevalent than the regulatory record would suggest.

Comparison of DDE and PCB

We have observed a herd that was simultaneously contaminated with high levels of DDE and Aroclor 1254 from a silo. By a fortuitous set of circumstances, the cows had been removed from the sources of both contaminations at about the same time. A detailed description of this work has been prepared (4).

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† In this paper the term PCB is interchangeable with Aroclor 1254 or, in the case of milk and body fat, residues derived from Aroclor 1254.
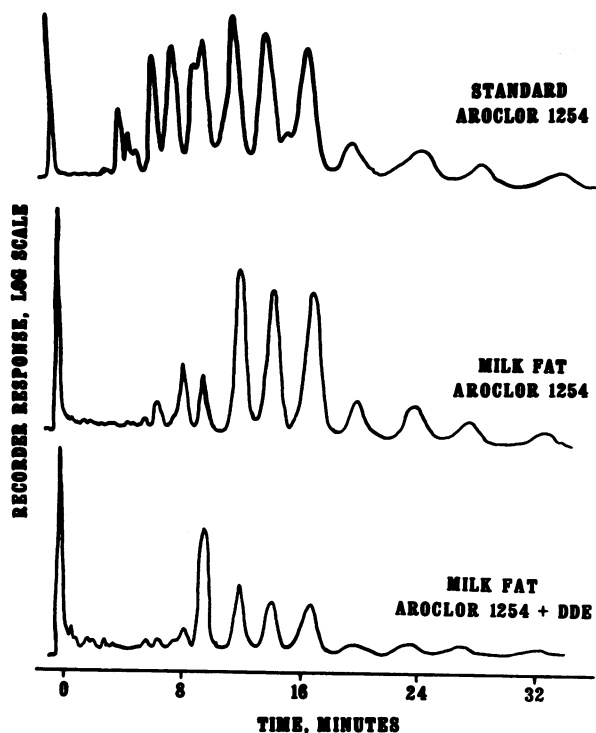
FIGURE 1. Gas chromatograms of Aroclor 1254 standard and residues in milk. The 6 ft. × ¼ in. o.d. column was packed with 10% DC-200 on Gaschrom-Q. A 95% argon–5% methane mixture was used as carrier gas and the column temperature was 200°C.

The average concentration of PCBs in milk fat was 19.3 μg/g when the silage feeding stopped. This concentration declined to 10.3 μg/g within three weeks. We continued to sample the individual cows at three-week intervals for approxi-

Table 1. PCB concentrations in milk fat from farms with contaminated silos[a]

Farm	11/10	12/7	1/21	3/24	5/11
			(μg/g)		
1	9.6	8.8	6.7	N.S.	7.0
2	7.5	5.7	6.7	6.6	*3.5*
3	N.S.[b]	*1.5*	7.4	5.0	*2.1*
4	N.S.	*2.0*	*1.4*	*1.4*	6.0
5	*3.5*	*2.7*	6.1	10.3	10.5
6	N.S.	*1.0*	6.2	5.2	*2.9*

[a] Silage was not being fed when italicized samples were obtained.
[b] Not Sampled.

Table 2. Average rates of decline of DDE and PCB concentration in milk fat of cows in various stages of lactation.[a]

Group	Cows	DDE	PCB
	(No.)	(day⁻¹)	(day⁻¹)
Mid lactation	16	0.0145	0.0147
Early lactation	9	0.0107	0.0106
Late lactation	6	0.0147	0.0140
All	31	0.0134	0.0134

[a] The values are for the constant, k, in the linear form of the first-order equation: $\ln C = \ln C_0 - kt$ where C is the milk fat concentration, C_0 is the initial concentration, and t is time in days. Fitted by least squares.

mately four months. After three weeks the decline in concentration was linear, when plotted on semilog graph paper, suggesting first order kinetics.

First-order rate constants were calculated for both DDE and PCBs for the individual cows. A summary is presented in Table 2. The overall average of the rate constants for PCB was identical to that for DDE. The average DDE rate constant was similar to the rate that we have previously reported (5). There was a tendency for cows in early lactation to have lower rate constants than the other groups of cows.
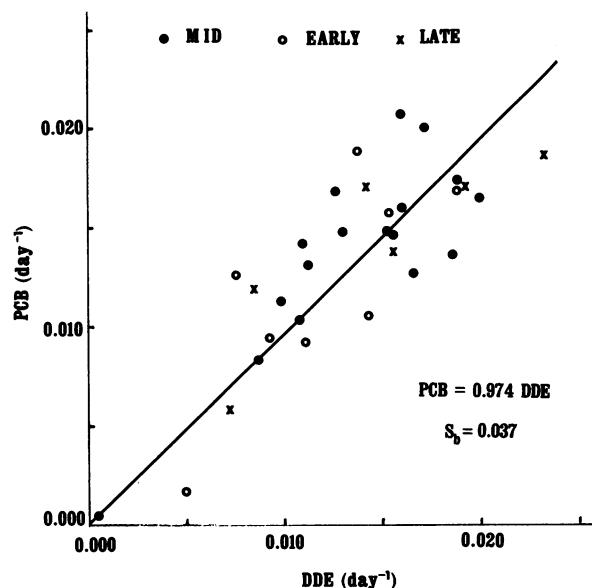

FIGURE 2. Relationship of the rates of decline of DDE and PCB concentration in milk fat of individual cow. Mid, early, and late are stages of lactation (4).

Table 3. Effect of phenobarbital and phenobarbital with activated carbon on the concentration of DDE and PCB in milk fat.[a]

| Treatment | Cows (No.) | Concentration | | Final Initial | S.D. |
		Initial (μg/g)	Final (μg/g)		
			DDE		
Control	5	11.6	5.2	0.45	±0.05
Pheno-barbital	10	9.5	4.3	0.45	±0.15
Pheno-barbital+ Carbon	6	8.0	3.7	0.46	±0.20
			PCB		
Control	5	19.3	7.4	0.38	±0.08
Pheno-barbital	10	17.7	7.0	0.40	±0.11
Pheno-barbital+ Carbon	6	22.3	8.5	0.37	±0.14

[a] Observation period was 6 weeks. Phenobarbital was fed at 5 grams per day for the first three weeks and activated carbon at one kilogram per day for the 6 weeks.
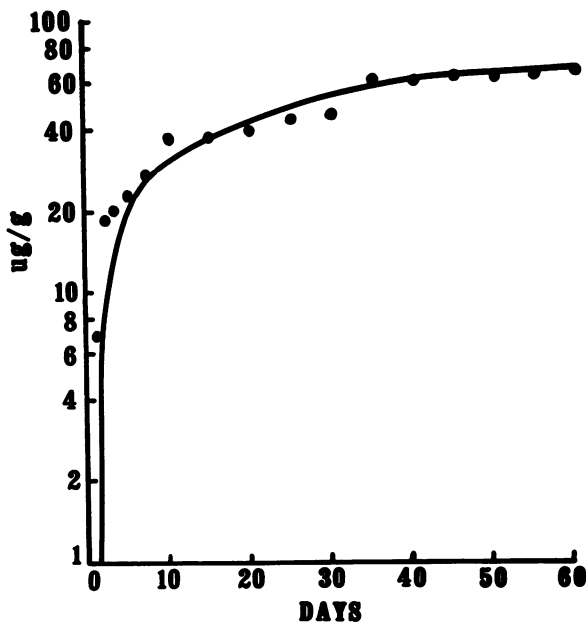
FIGURE 3. Concentration of PCB in milk fat of cows fed 200 mg/day Aroclor 1254 for 60 days. Each point is an average of nine cows.
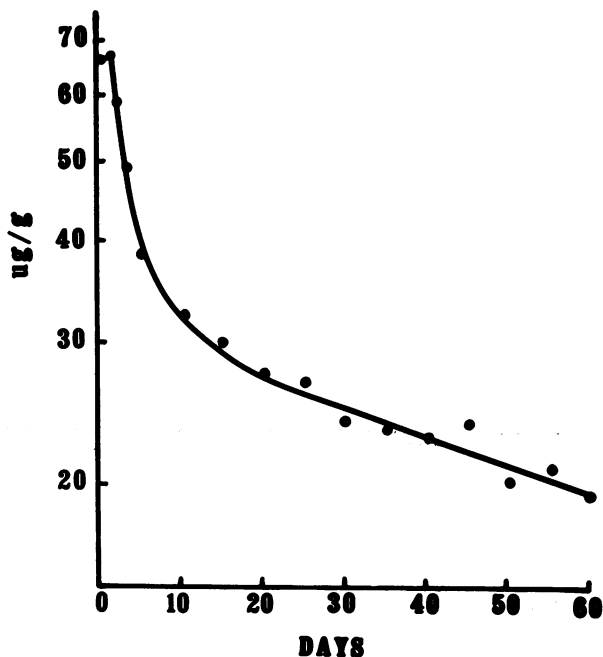
FIGURE 4. Concentration of PCB in milk fat after feeding Aroclor 1254 stopped. Each point is an average of nine cows.

Relationship of the rate constants for DDE and PCBs within individual cows is presented in Fig. 2. The correlation (r = 0.82) between the rate constants of the two compounds was significant ($P<0.01$). The usual linear regression was calculated, and the intercept did not differ significantly from zero (6). Therefore, the regression in Fig. 2 was recalculated, forcing the intercept through zero. The regression coefficient, 0.974, was essentially unity.

In some cases there was considerable variation between the rate constants for DDE and for

Table 4. Concentration of PCB in milk fat and body fat of cows fed 200 mg/day PCB for 60 days.

Time	Milk fat	Body fat
(days)	(μg/g)	(μg/g)
30	45.6±8.4[a]	21.9±8.5
60	66.7±8.4	44.0±12.2
90	24.0±3.0	40.3±6.4
120	19.4±2.4	28.8±6.2

[a] Standard deviation.

Table 5. Normalized concentration of chlorinated hydrocarbon compounds in milk fat and body fat of cows after continuous feeding.[a]

Compound	Reference	Observations (No.)	Milk fat 20 days (μg/g)	Milk fat 60 days (μg/g)	Body fat 60 days (μg/g)
Aroclor 1254		9	0.20	0.33	0.20
DDE	(5)	3	0.21	0.29	0.23
	(7, 8)	12	0.27	—	—
Dieldrin	(7, 8)	12	0.19	—	—
DDD	(5)	3	0.063	0.077	0.046
p,p'-DDT[b]	(5)	3	0.036	0.049	0.033
	(7, 8)	12	0.036	—	—
	(9)	4	0.106	—	—
o,p'-DDT[b]	(9)	4	0.023	—	—

[a] Values are normalized to an intake of 1 mg/day.
[b] Values are sum of DDD and DDT.

PCBs within a given cow. The rate constants for the two compounds within a given cow were tested statistically (6), but the differences were not significant.

The usefulness of phenobarbital alone, or in combination with activated carbon, in accelerating the reduction in milk concentration of DDE and PCBs was tested using three groups of cows. Neither phenobarbital nor phenobarbital in combination with activated carbon had an effect on the relative reduction in concentration of either compound (Table 3). We have previously reported a small effect of phenobarbital on DDE, but we did not consider it of practical significance (7). The absence of an effect here is consistent with that interpretation. The failure of activated carbon, even in combination with phenobarbital, to affect the reduction of DDE and PCB concentrations is consistent with our previous interpretation of laboratory experiments (8).

Aroclor 1254 Feeding Study

We have carried out a controlled feeding study using nine Holstein cows. The cows were fed 200 mg per day Aroclor 1254 for 60 days. Milk samples were obtained periodically during the 60-day feeding period and the subsequent 60-day period. Body fat samples were obtained by biopsy at 30-day intervals.

The cows were selected to have a range in both the stage of lactation and level of milk production.

However, preliminary examination of the results indicates that there was little difference in the residue levels due to these factors. Therefore, only average data for all cows are presented.

The average level of PCBs in the milk fat during the feeding period is presented in Fig. 3. The standard deviation of any given point did not exceed $\pm 10\%$ of the mean. The concentration of PCBs increased rapidly and approached a "steady state" at about 40 to 60 days. As expected from our field studies, the shape of the curve is similar to that for other chlorinated hydrocarbon pesticides, particularly DDE (5).

The decline in PCB concentration in the milk for the 60 days after feeding stopped is presented in Fig. 4. This curve is quite typical of previous findings with DDE and was resolved into a two-component first-order system (5). The equation for the curve normalized to an initial concentration of 1.0 μg/g is:

$$C = 0.52e^{-0.25t} + 0.48e^{-0.008t}$$

Where C is concentration, e is the base of the natural logarithms, and t is days.

The constants are similar to our previous observations for DDE (5). While the rate for the second component in this study was less than the values found in the field, it was within the range of values that one might expect.

The level of PCB in body fat at various times is presented in Table 4. For reference purposes, milk fat levels at these times are also presented.

While the PCBs were being fed, the level in milk fat was higher than that in body fat. When feeding stopped, milk fat levels dropped below and appear to reflect body fat levels.

We have conducted a number of studies with chlorinated hydrocarbon pesticides under conditions similar to those of this PCBs study. A summary of levels in milk fat at 20 and 60 days and body fat at 60 days with normalized intakes is presented in Table 5. The levels of PCBs are similar to levels of DDE and dieldrin in corresponding samples. In contrast DDD, p,p'-DDT, and o,p'-DDT are transferred to the body fat and milk fat at much lower rates than DDE and PCBs.

Conclusions

Farms having silos that have been treated with PCB-containing paints will frequently have residues of PCBs in milk which exceed the FDA guidelines. The behavior of this residue in the cows is similar to DDE and other chlorinated hydrocarbon pesticides resistant to metabolic degradation. The only practical countermeasures appear to be to decontaminate the silos sufficiently to remove most of the PCBs or to discontinue the use of these silos completely. Limited field experience and the similarity of PCBs to DDE suggest that there are no practical procedures to minimize the transfer of PCBs from the diet to the milk or to speed up the elimination of PCBs from cows.

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